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# Pyrethroid Illnesses in California, 1996-2002

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## I. Introduction

With regulatory limitations on the use of cholinesterase-inhibiting insecticides, synthetic pyrethroids have become increasingly important in both agricultural and structural pest control (USEPA 1996, 2000). Pyrethroid chemistry and action are classified as type I or type II depending on the alcohol substituent. Type I pyrethroids contain a descyano-3-phenoxybenzyl or other alcohols. The type II, or alpha-cyano pyrethroids, contain α-cyano-3-phenoxybenzyl alcohol, which increases insecticidal activity about 10-fold.

The California Department of Pesticide Regulation (DPR), Worker Health and Safety Branch (WH&S) is responsible for public and worker safety wherever pesticides are used in the state. Their mission is accomplished through several programs: exposure monitoring, exposure assessment and mitigation, pesticide illness surveillance, and workplace evaluation and industrial hygiene. The WH&S Pesticide Illness Surveillance Program (PISP) is generally acknowledged as the nation's most compre-

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hensive and reliable reporting system (Mehler et al. 1992; U.S. Government Accounting Office 1994). Data generated from illness reports have made DPR's worker protection program a model for other states (Western Farm Press 2001). WH&S has previously analyzed PISP data to develop retrospectives of illnesses, injuries, and deaths related to pesticide exposures in California to determine risk factors for exposure to organophosphate insecticides in agricultural workers and to summarize investigations of exposure incidents related to specific pesticides (Maddy et al. 1990; O'Malley et al. 1990; O'Malley 1998b; O'Malley and Verder-Carlos 2001). WH&S has also evaluated PISP data to determine the effectiveness of California's worker protection program regarding field posting, hazard communication, notification and retaliation requirements, and irrigator and pesticide handler exposures (Fong 2001; McCarthy 2003; Spencer 2001).

This survey summarizes California's recent experience with pyrethroid-related illnesses, using PISP data, pesticide use reporting data, and investigations of three large group illness episodes related to exposure to type II pyrethroids cyfluthrin and λ-cyhalothrin (Edmiston et al. 1998, 1999). Cases were reviewed by use, structural class and routes of exposure for the years 1996-2002, focusing on 317 cases involving exposure to one or more pyrethroid compounds (DPR, unpublished data).

Illnesses associated with pyrethroid exposures during packaging, during agricultural and indoor/structural applications, and resulting from indoor residue exposures have been documented (He et al. 1988; Muller-Mohnssen 1999; Pauluhn 1996; Prohl et al. 1997). Few studies have evaluated agricultural occupational exposures to pyrethroid residues (Kolmodin-Hedman et al. 1982, 1995). Although pyrethroid residues on crops, agricultural products, and foliage have previously been investigated, pyrethroid dissipation is not well understood, as compared to the dissipation of organophosphorus pesticides (Argauer et al. 1997; Bellows et al. 1993; Dejonckheere et al. 1982; Edmiston et al. 1999; Estesen and Buck 1990; Giles et al. 1992; Hernandez et al. 1998; McEwen et al. 1986; Miyata et al. 1993; Nakamura et al. 1993; Papadopoulou-Mourkidou et al. 1989). This review provides recent data on agricultural and nonagricultural illnesses related to pyrethroid exposures and augments the data available on pyrethroid residue dissipation.

## II. Pyrethroid Mode of Action

Pyrethroids cause prolongation of sodium channel currents in the nervous system, and many of their adverse effects are related to this property. For all pyrethroids, the major neurotoxic hazard is acute excitation. Exposure to pyrethroids can cause dermal irritation and paresthesia, very often without producing visible erythema of the skin (Cagen et al. 1984). Similar irritant effects occur in the respiratory tract (Pauluhn et al. 1996; Pauluhn 1999; Ray 2000). Systemic toxicity has been reported on ingestion

(O'Malley 1997) and may also occur with a high degree of occupational exposure (He et al. 1988,1989). The α-cyano subgroup of compounds (type II pyrethroids) produce more prolonged sodium channel currents than type I compounds and often are relatively more potent as insecticides and mammalian toxins (Casida et al. 1983). In laboratory animals, the toxidromes produced by type I and type II compounds are considered distinct (Ray 2000). Type I pyrethroids cause the simplest poisoning syndrome, characterized in animal studies by severe fine tremor, marked reflex hyperexcitability, sympathetic activation, and, for dermal exposure, paresthesia. Type II pyrethroid poisoning is more complex and causes more severe symptoms. In addition to sympathetic activation and paresthesia, the toxidrome exhibited profuse watery salivation, coarse tremor, increased extensor tone, moderate reflex hyperexcitability, choreoathetosis, and seizures (Ray 2000).

## III. Illness, Exposure, and Pesticide Use Data

### A. Illness and Use Report Data

California has implemented a full-use reporting system for pesticides since 1990. Since 1981, DPR's PISP has maintained a database of pesticide-related illnesses and injuries. DPR receives case reports from physicians and via Workers' Compensation records. The local county agricultural commissioner investigates circumstances of exposure for each case. WH&S then evaluates the medical records and investigative findings and enters the data into an illness registry. WH&S may conduct investigations when it appears that significant hazards contributed to the exposures, such as in the three group episodes of exposure to type II pyrethroids discussed later.

Table 1 summarizes both use and illness data associated with the 317 pyrethroid-related illnesses reported to PISP between 1996 and 2002. For the 13 active ingredients associated with those illnesses, 4,629,851 lb (2,100,068 kg) were reported to DPR as being used in the state during the same time period (Table 1) (DPR 2004). Two compounds, permethrin and cypermethrin, accounted for 75% of total use. Type II compounds accounted for 42.8% of the reported pounds used (1,979,352lb; 897,820 kg), but were associated with 220 (69.4%) of the cases. A single compound, cyfluthrin, was associated with 122 cases (55% of illnesses related to type II pyrethroids and 38.4% of all pyrethroid illnesses). Cyfluthrin use (308,191 lb; 139,793 kg) comprised 15.6% of total type II usage and 6.6% of total pyrethroid usage. Type I compounds accounted for 2,650,5001b (1,202,248 kg), 57.2% of the reported use, and 97 (30.6%) of the reported illness cases. The type I compounds most frequently associated with illnesses were resmethrin (38 cases) and permethrin (44 cases).

Agricultural use includes uses for treatment of crops, nurseries, livestock, agricultural research facilities, and the handling of raw agricultural commodities in packing houses. These uses accounted for 118 (37.3%) of the

Table 1. Summary of Pyrethroid-Related Illnesses[a] and Reported Pyrethroid Use[b] in California, 1996-2002.

| Pyrethroid | No. illnesses | Reported use (lb) | Reported use (kg) |
|---|---|---|---|
| Type I | | | |
| Allethrin | 3 | 571 | 259 |
| Bifenthrin | 11 | 199,302 | 90,402 |
| Permethrin | 44 | 2,445,525 | 1,109,273 |
| Phenothrin | 1 | 583 | 264 |
| Resmethrin | 38 | 4,519 | 2,050 |
| Total Type 1 | 97 | 2,650,500 | 1,202,248 |
| Type II | | | |
| Cyfluthrin | 122 | 308,191 | 139,793 |
| Cypermethrin | 43 | 1,091,419 | 495,060 |
| Deltamethrin | 3 | 45,216 | 20,510 |
| Esfenvalerate | 26 | 234,886 | 106,543 |
| Fenpropathrin | 2 | 117,637 | 53,359 |
| Fluvalinate | 3 | 19,929 | 9,040 |
| λ-Cyhalothrin | 12 | 153,621 | 69,681 |
| Tralomethrin | 9 | 8,453 | 3,834 |
| Total Type II | 220 | 1,979,352 | 897,820 |
| Total Use | | 4,629,852 | 2,100,068 |

[a]Reported to California's Pesticide Illness Surveillance Program; illness was related to one or more pyrethroids as the primary causal agent.

[b]Reporting required for agricultural use, all use by pest control businesses, all restricted materials, all institutional use of pesticides on the groundwater protection list, and all post-harvest commodity treatment; does not include home and garden use of products sold overthe-counter.

reported cases, with all but 2 cases (related to bystander drift) associated with employment. Agricultural cases were primarily (95%) associated with applications to either crops (86 illnesses, 62 of which were in oranges) or food processing/storage facilities (26 illnesses). For the 199 cases (62.8%) associated with nonagricultural use, 132 (66.3%) were occupationally related. Nonagricultural cases were predominantly (80%) related to applications inside or outside of buildings and homes (159 illnesses). Nonagricultural uses included residential, retail, service and institutional uses, structural pest control, rights-of-way, parks, landscaped urban areas, and public health vector control (Table 2).

## B. Group Versus Individual Illness

Approximately equal numbers of illnesses resulted from individual exposures (167 cases) and group exposures (150 cases). The majority of illnesses related to agricultural use (75%) occurred in seven group episodes (88 ill-

Table 2. Summary of Pyrethroid-Related Illnesses Reported to California's Pesticide Illness Surveillance Program, 1996-2002, by Number of Persons Involved in Group Illness Episodes[a] versus Individual Illnesses.[b]

| Year | No. persons exposed in group illness episodes[a] (no. episodes) | | No. persons exposed in individual illnesses[b] | | Total |
|---|---|---|---|---|---|
| | Agricultural[c] | Nonagricultural[d] | Agricultural[a] | Nonagricultural[a] | |
| 1996 | 2 (l) | 6 (l) | 1 | 16 | 25 (2) |
| 1997 | 55 (2) | 10 (5) | 9 | 22 | 96 (7) |
| 1998 | 0 | 7 (1) | 4 | 7 | 18 (l) |
| 1999 | 0 | 21 (3) | 4 | 15 | 40 (3) |
| 2000 | 26 (2) | 9 (4) | 4 | 25 | 64 (6) |
| 2001 | 5 (2) | 9 (2) | 4 | 17 | 35 (4) |
| 2002 | 0 | 0 | 4 | 35 | 39 (0) |
| Total | 88 (7) | 62 (16) | 30 | 137 | 317 (23) |

[a]Episode: a single exposure event; one or more persons may became ill from a single pesticide exposure incident.

[b]Illness: indicates a single individual became ill.

[c]Uses intended to contribute to the production of agricultural commodities including crops, nursery products, and livestock. It includes transportation and storage of pesticides on farmlands and use at agricultural research facilities and in packing houses. It excludes forestry operations and the manufacture, trans-portation, and storage of pesticides before arrival at the site of agricultural production. Pesticides used agriculturally retain their agricultural designation regardless of exposure location.

[d]The pesticide(s) were not intended to contribute to the production of agricultural commodities. This includes residential, retail, service, and institutional uses, structural pest control, use in rights-of-way, parks, and landscaped urban areas, and the manufacture, transportation, and storage of pesticides except on farmlands.

Table 3. Summary of Symptoms for Pyrethroid-Related Illnesses Reported to California's Pesticide Illness Surveillance Program, 1996-2002.

| Symptom array | Type I pyrethroids | Type II pyrethroids | Total illnesses by symptom array |
|---|---|---|---|
| Irritant[a] | 26 | 107 | 133 |
| Irritant/systemic | 41 | 95 | 136 |
| Systemic | 30 | 18 | 48 |
| Total illnesses | 97 | 220 | 317 |

[a]Dermal, eye, and/or respiratory irritant symptoms.

nesses). The majority of nonagricultural use-related illnesses (69%) were due to individual exposures (137 illnesses).

### C. Symptom Array and Exposure Route

The symptom arrays associated with the pyrethroid illnesses are summarized in Table 3. Irritant effects or paresthesias of the eye, skin, or respiratory tract were present in 269 cases (84.9%). Type II pyrethroids were more frequently associated with isolated irritant symptoms (107 cases) than the type I pyrethroids (26 of 97 cases). Systemic symptoms were reported in 184 illnesses (58% of cases). Isolated systemic effects occurred in 48 cases (15.1%), but systemic effects were also present in 136 (50.6%) of the 269 cases with irritant symptoms. Isolated systemic symptoms were reported in 30 of 97 cases related to type I pyrethroids and in 18 of 220 cases involving type II pyrethroids. Specific symptoms reported included headache, nausea, vomiting, epigastric pain, weakness, lethargy, fatigue, dizziness, sweating, and/or muscle pain.

Residue exposures accounted for 158 illnesses (49.8%), with the majority of the illnesses (149, 94%) occurring in the following activity categories: field worker (69 cases), routine indoor exposures (55 cases), and packing/processing (25 cases) (Table 4). Direct exposures accounted for 75 illnesses (23.7%), with the majority occurring while persons were involved in application (39 cases) and mixing/loading (9 cases) tasks, emergency response activities (8 cases), and activities categorized as other (8 cases). Drift exposures accounted for 62 illnesses (19.6%), with the majority occurring during routine indoor activities (34 cases), application tasks (15 cases), and routine outdoor activities (9 cases). Because of the three illness clusters, the residue exposure cases accounted for a large majority (79.7%) of the 118 agricultural cases. Only 32.1 % of the 199 nonagricultural cases were associated with residue exposure. Seven illnesses resulted from intentional or accidental ingestion and 15 had unknown/other exposure routes. The latter group included notable illnesses or injuries following explosions that occurred when excessive application of indoor foggers ignited vapors from pilot lights.

### D. Pesticide Use Violations

Single or multiple violations of pesticide use regulations contributed to exposures in 90 of the 317 illnesses (28.4%); 76 were related to nonagricultural pyrethroid use. The most common violations involved pesticide misuse (78 cases), including failure to control drift, failure to notify building occupants of a structural application, and use of higher than label rates in applications (including residents using multiple aerosol foggers to treat dwellings). Other contributing violations, alone or in combination with pesticide misuse, included failure to wear proper protective equipment and early reentry following an application.

## IV. Three Group Illness Episode Investigations

The following summarizes WH&S investigations of three group illness episodes where respiratory and dermal irritation symptoms were prominent. The episodes were related to agricultural exposures to type II

Table 4. Summary of Pyrethroid-Related Illnesses Reported to California's Pesticide Illness Surveillance Program, 1996-2002, by Symptom Array, Exposure Route, and the Relationship to Agricultural[a] or Nonagricultural[b] Pesticide Use.

| | | Symptom array | | |
|---|---|---|---|---|
| Exposure route | Total | Irritant[c] | Irritant/systemic | Systemic |
| Direct[d] | 75 | 45 | 18 | 12 |
| Agricultural[a] | 11 | 10 | 0 | 1 |
| Nonagricultural[b] | 64 | 35 | 18 | 11 |
| Drift[e] | 62 | 20 | 30 | 12 |
| Agricultural | 12 | 4 | 4 | 4 |
| Nonagricultural | 50 | 16 | 26 | 8 |
| Residue[f] | 158 | 57 | 81 | 20 |
| Agricultural | 94 | 31 | 50 | 13 |
| Nonagricultural | 64 | 26 | 31 | 7 |
| Ingestion[g] | 7 | 0 | 4 | 3 |
| Unknown/other | 15 | 11 | 3 | 1 |
| Agricultural | 1 | 0 | 0 | 1 |
| Nonagricultural | 14 | 11 | 3 | 0 |
| Grand totals | 317 | 133 | 136 | 48 |

[a]Uses intended to contribute to the production of agricultural commodities including crops, nursery products, and livestock. It includes transportation and storage of pesticides on farmlands and use at agricultural research facilities and in packing houses. It excludes forestry operations and the manufacture, transportation, and storage of pesticides before arrival at the site of agricultural production. Pesticides used agriculturally retain their agricultural designation regardless of exposure location.

[b]The pesticide(s) were not intended to contribute to the production of agricultural commodities. This includes residential, retail, service, and institutional uses, structural pest control, use in rights-of-way, parks, and landscaped urban areas, and the manufacture, transportation, and storage of pesticides except on farmlands.

[c]Dermal, eye, and/or respiratory irritant symptoms.

[d]Body contact with appreciable quantities of pesticide.

[e]Airborne exposure during pesticide application or preparation for application.

[f]Exposure to any amount or component of a pesticide that remains in the environment after application.

[g]Accidental or intentional ingestions.

pyrethroids cyfluthrin (two episodes) and λ-cyhalothrin (one episode) (DPR, unpublished data; Edmiston et al. 1998,1999). The cyfluthrin illnesses were included among the 317 discussed previously. The λ-cyhalothrin illnesses were not included in the preceding discussion because propargite and sulfur residues were also present. Although all three episodes involved harvesters entering treated fields well after applicable restricted entry intervals had elapsed, the product in the λ-cyhalothrin episode had been mistakenly and illegally applied to grapes, at rates far higher than legally permitted for registered crop uses.

## A. Cyfluthrin Orange Harvester Episodes

Cyfluthrin was involved in 121 of the 317 illnesses described previously. Sixty-nine illnesses were related to agricultural uses, with 61 illnesses occurring in oranges: the two group episodes described below involved 55 harvesters. The other 6 illnesses included three field workers exposed in individual illnesses, two applicators and one mixer/loader. Fifty-two illnesses occurred in nonagricultural use settings, predominantly (38 illnesses) affecting persons exposed to residue (29 illnesses) or drift (9 illnesses) while involved in routine indoor activities.

The two group illness episodes occurred in Tulare County in May 1997, when 55 Valencia orange harvesters became ill while working in groves that were treated with cyfluthrin 3-10d earlier (Edmiston et al. 1998, 1999). Although the restricted entry interval was 12 hr, the registration status of cyfluthrin changed in April 1997 when the preharvest interval was reduced from 150d to the day of harvest. The harvesters sought medical care for symptoms involving primarily respiratory irritation, including rhinitis (54.2%), sneezing (81.9%), coughing (21.7%), and sore throat (33.7%). Other symptoms included headache (21.7%), nausea (6.0%), and skin (16.9%) and eye irritation (16.9%). The symptoms were transitory, and most of the harvesters returned to work the following day.

## B. Cyfluthrin Inhalation Monitoring Study

As part of the episode investigation, WH&S monitored the inhalation exposures of three experienced orange harvesters during 6 hr of harvesting (Edmiston et al. 1998). The monitoring began 30hr after the Valencia orange grove was treated with cyfluthrin at the rate of l lb/A (0.45 kg/A). Cyfluthrin dust was trapped on 25-mm glass fiber filters housed in Institute of Occupational Medicine (IOM) samplers attached via tubing to personal air sampling pumps. Table 5 presents cyfluthrin residues found on the filters (mean = 5.13 μg/sample), inhalation concentration (mean = 7.13 μg/m$^3$),

Table 5. Potential Inhalation Exposure for Orange Harvesters Exposed to Cyfluthrin Residues in California, 1997.

| | Cyfluthrin residues (μg/sample) | Inhalation concentration (μg/m$^3$) | Potential inhalations (μg/kg) | Absorbed dosages (μg/kg/day) |
|---|---|---|---|---|
| Worker 1 | 2.19 | 3.04 | 38.72 | 0.553 |
| Worker 2 | 5.60 | 7.78 | 100.24 | 1.432 |
| Worker 3 | 7.60 | 10.56 | 136.42 | 1.949 |
| Mean | 5.13 | 7.13 | 91.79 | 1.311 |

'Based on 8-hr exposure.

potential inhalation exposure (mean = 91.79 µg/kg, based on 8-hr exposure), and estimated absorbed dosage (mean = 1.311 µg/kg/d).

Numerous cyfluthrin toxicology studies on file with DPR show irritant symptoms associated with a peak concentration of 100 $\mu g/m^3$; this is also reported in the literature (Pauluhn and Machemer 1998). Adequate data to determine an acute no observable adverse effect level (NOEL) for human respiratory irritation following continuous exposures are not available. However, because the monitoring study found average inhalation concentrations provided a margin of exposure (MOE) of less than 100 fold compared to the experimentally established irritant threshold, DPR placed all products containing cyfluthrin into reevaluation in May 1998 (DPR, unpublished data). As part of the reevaluation process, DPR has required the primary registrant of cyfluthrin products to conduct and submit the results of inhalation irritation threshold studies and monitor corn harvesters exposed to cyfluthrin residues.

### C. Cyfluthrin Dislodgeable Foliar Residue (DFR) Monitoring

WH&S sampled DFR from the episode groves and the grove where the inhalation monitoring study was conducted to determine average cyfluthrin residues during the monitoring period (Edmiston et al. 1999). To evaluate residue dissipation, WH&S sampled seven Valencia orange groves located within 50 mi of the episode groves over a 9-wk period following application. Pest control operators treated all sampled groves with cyfluthrin at 0.101b (0.05 kg) active ingredient/A in 100-250 gal (379-946L) water. All tank mixes also included nutrients or buffers.

The following average cyfluthrin residues were found for the episode groves (6-12 d postapplication) and the grove where the inhalation monitoring study was conducted (11 d postapplication):

Episode groves, $0.039 \pm 0.011\ \mu g/cm^2$

Monitoring groves, $0.035 \pm 0.007\ \mu g/cm^2$

Using nonlinear regression analysis, the model $In\ \mathrm{DFR} = a \pm b^{*} \sqrt{(days)}$ was the lowest order model that fit well for most of the fields. Table 6 displays the associated data for intercept, slope, and $R^2$ under this model. Initial deposition (as reflected by the model intercept) was similar across groves. The dissipation rates in the seven groves fell into two distinct decay patterns, with more rapid decay in groves 1-4 (overall average half-life = 4.9 d) and a considerably longer decay in groves 5-7. The half-life for groves exhibiting slower residue dissipation under the fitted model is not constant, as it would be with a first-order exponential decay model. Instead, each half-life is longer than the preceding one. The first two half-lives for groves 5-7 can be approximated as 11 and 32 d, respectively. In these groves, approximately 10%-20% of the initial residue was still present at 65 d

Table 6. Cyfluthrin Dislodge able Foliar Residue (DFR) Dissipation in eight Valencia Orange Groves Treated with Cyfluthrin in California, 1997.[a]

| Grove | Intercept | Slope | Average half-life (d) | $R^2$ |
|---|---|---|---|---|
| 1 | -3.009 | -0.397 | 5.73 | 0.63 |
| 2 | -2.625 | -0.441 | 4.82 | 0.81 |
| 3 | -2.969 | -0.450 | 4.67 | 0.75 |
| 4 | -2.802 | -0.470 | 4.37 | 0.89 |
| 5 | -2.754 | -0.233 | 19.6 | 0.67 |
| 6 | -2.782 | -0.258 | 14.7 | 0.69 |
| 7 | -3.050 | -0.152 | 95.6 | 0.55 |

'Fit of the model *In DFR* $= a + b * \sqrt{(days)}$ *(days after application).*

*Source:* Edmiston et al. (1999).

postapplication. Cyfluthrin appears to exhibit a variable dissipation rate, with the use of urea and/or potassium nitrate possibly associated with slower dissipation.

### D. λ-Cyhalothrin Illness Episode

λ-Cyhalothrin, first registered in California in 1998, was the causal pyrethroid in 12 of the 317 illnesses previously discussed. Symptom arrays included irritant (5 cases), irritant/systemic (6 cases), and systemic (1 case). Six illnesses resulted from exposure to residues. Although the λ-cyhalothrin technical fact sheet states that adverse health effects include both skin and respiratory irritation, it documents only skin irritation as an adverse effect (National Pesticide Telecommunication Network 2004). The incidents cited involved laboratory personnel and workers who either handled concentrated λ-cyhalothrin and/or applied dilute λ-cyhalothrin solutions. The following describes an illness episode in which field workers developed acute respiratory symptoms on exposure to extremely high foliar residues of λ-cyhalothrin.

In September 1999, 11 raisin harvesters developed acute respiratory irritation symptoms when they entered a Fresno County vineyard (episode field) and were exposed to residues of λ-cyhalothrin, propargite, and sulfur (unpublished data, DPR). Within approximately 3 hr, crew members experienced sneezing, flu-like symptoms, and burning and itching on their arms, neck, face, and eyes. Seven workers received medical treatment. Subsequent investigation confirmed that the pesticide product Warrior Insecticide (U.S. EPA registration number 10182-00434-AA, containing 13.1% λ-cyhalothrin; Syngenta Crop Protection, Inc.), which was not registered for use on grapes, was mistakenly mixed and applied 45 d prior at 35 times the highest legal rate for any crop (Syngenta Crop Protection Inc. 2004). Gas chromatography analyses of eight DFR samples verified mean residues of λ-cyhalothrin ($0.43 \pm 0.10$ μg/cm$^2$), propargite ($0.35 \pm 0.11$ μg/cm$^2$), and sulfur (0.31 ± 0.28 μg/cm$^2$) on the grape leaves. The Fresno County Agricultural Commissioner declared the episode field a hazardous area due to high propargite and λ-cyhalothrin residues and prohibited worker reentry. WH&S compared episode DFR with 1998 λ-cyhalothrin residues for romaine lettuce and found episode field residues were approximately 12 times greater than those measured previously (Hernandez et al. 1998). The effects of exposure to average λ-cyhalothrin DFR levels of 0.43 μg/cm$^2$ have not been previously documented (Extension Toxicology Network 2004).

All eight DFR samples from the episode field showed propargite levels above 0.20 μg/cm$^2$, the estimated safe reentry level for repeated exposures to nectarine harvesters (O'Malley et al. 1990). Propargite and sulfur have been implicated in a number of dermal illness episodes over the past 20 yr (Gammon et al. 2001; Maddy et al. 1981; O'Malley et al. 1990; O'Malley 1998a; Winter and Kurtz 1985). Although propargite levels may have had a role in the incident, WH&S considered the high levels of λ-cyhalothrin to be the primary cause. Because information was unavailable for estimating the decay rate for such high residues, WH&S remediation for the episode field included posting the field against worker entry until natural defoliation had occurred and specifying that others who may enter before defoliation wear a properly fitted N95 particulate respirator, a disposable coverall, boots, and disposable gloves (DPR, unpublished data). The applicator was cited and paid a civil penalty of $1,000. λ-Cyhalothrin exposure resulting in illness was noted in a previous indoor applicator study, in which applications were made at legal rates (Moretto 1991).

## V. Conclusions

In limited DPR monitoring studies associated with harvesting oranges at approximately two-thirds the normal work intensity, cyfluthrin air levels (10 μg/m$^3$ cyfluthrin, measured as a time-weighted average) approached experimentally established irritant thresholds (LOELs) for airborne cyfluthrin (100 μg/m$^3$ measured as an initial peak level) (Edmiston et al. 1998; Pauluhn and Machemer 1998). These observations and the common set of symptoms seen in experimental studies, PISP illnesses, and agricultural, structural, and indoor residue exposures reported in the literature suggest that field residues can cause irritant respiratory symptoms. It is unclear whether nonspecific symptoms such as nausea and headache reported in some of those indicate true systemic toxicity. Additional data are needed to establish threshold levels for both irritant and systemic symptoms for cyfluthrin and other pyrethroids.

For regulations based upon reentry intervals or waiting periods, additional dissipation data may also be necessary. Pyrethroid residues on crops, agricultural products, and foliage have previously been investigated, but pyrethroid dissipation is not well understood, as compared to the dissipation of organophosphorus pesticides (Argauer et al. 1997; Bellows et al. 1993; Dejonckheere et al. 1982; Edmiston et al. 1999; Estesen and Buck 1990; Giles et al. 1992; Hernandez et al. 1998; McEwen et al. 1986; Miyata et al. 1993; Nakamura et al. 1993; Papadopoulou-Mourkidou et al. 1989). Dissipation appears to vary significantly among pyrethroids and between crops, which hampers developing models to describe residue transfer in occupational settings. Previous studies of pyrethroid residues on cotton and orange foliage found half-lives of 5.3 and 6.2 d, respectively (Bellows et al. 1993; Estesen and Buck 1990). This result is similar to the dissipation exhibited in groves 1-4 in the WH&S cyfluthrin DFR study (see Table 6). Another study reported pyrethroid dissipation half-lives of 6.9-18.2 d for greenhouse-grown chrysanthemums, similar to the slower dissipation rate observed for groves 5 and 6 in the WH&S cyfluthrin DFR study (Giles et al. 1992). Dissipation as slow as that observed for grove 7 in WH&S' cyfluthrin DFR study (overall average half-life = 96 d; see Table 6) has not been reported in the literature. Dissipation following structural applications has been studied to only a limited extent, but sampling of carpet from treated homes in Germany has demonstrated that residues can persist for several years after application (Prohl et al. 1997). The Prohl study found that 83% of the subjects who had carpets removed had complete or partial improvement in nonspecific systemic and topical irritant symptoms compared to 16% of subjects who left treated carpet in place. These data are impressive even though the observational design of the study did not control for possible placebo effects of removing the treated carpet.

## Summary

This survey summarizes California's recent experience with illnesses related to pyrethroid exposures and augments the data available on pyrethroid inhalation exposure and residue dissipation. We reviewed California Department of Pesticide Regulation (DPR) Pesticide Illness Surveillance Program (PISP) data and DPR Pesticide Use Reporting (PUR) data for 13 pyrethroids used during 1996-2002 and identified 317 illnesses associated with exposure. PUR found a total of 4,629,852 pounds (2,100,068 kg) of the 13 active ingredients were applied during the 7 yr. Type II pyrethroids accounted for 1,979,352 (897,820kg) and 42.7% of the total pounds applied and 220 (69.6%) of the reported illnesses. Cyfluthrin was associated with 122 cases (55% of illnesses related to type II pyrethroids and 38.4% of all pyrethroid illnesses).

Agricultural uses accounted for 118 (37.3%) of the reported illness cases, with 116 cases associated with employment. For the 199 cases (62.8%) associated with nonagricultural use, 132 (66.3%) were occupationally related. Overall, approximately equal numbers of illnesses resulted from individual exposures (167 cases) and group exposures (150 cases). The symptom arrays associated with the pyrethroid illnesses included irritant effects or pares-

thesias of the eye, skin, or respiratory tract in 269 cases (84.9%). Type II pyrethroids were more frequently associated with isolated irritant symptoms (107 cases) than the type I pyrethroids (26 of 97 cases). Systemic symptoms were reported in 184 illnesses (58% of cases). Isolated systemic effects occurred in 48 cases (15.1%), but systemic effects were also present in 136 (50.6%) of the 269 cases with irritant symptoms. Residue exposures accounted for 158 illnesses (49.8%). Single or multiple violations of pesticide use regulations contributed to exposures in 90 of the 317 illnesses (28.4%); 76 were related to nonagricultural pyrethroid use.

We also report results of DPR Worker Health and Safety Branch (WH&S) investigations of three large group illness episodes related to exposure to type II pyrethroids cyfluthrin and λ-cyhalothrin that involved primarily respiratory irritation symptoms. An inhalation monitoring study found cyfluthrin air levels that approached experimentally established irritant thresholds for airborne cyfluthrin, from which a mean estimated absorbed dosage of 1.311 µg/kg/d was calculated. Although additional data are needed to establish threshold levels for both irritant and systemic symptoms for cyfluthrin and other pyrethroids, these observations suggest that field residues can cause irritant respiratory symptoms. DPR conducted a residue dissipation study in seven orange groves and estimated cyfluthrin residue half-lives. The dissipation rates fell into two distinct decay patterns, with more rapid decay in groves 1-4 (overall average half-life = 4.9 d) and a considerably longer decay in groves 5-7. The half-life for groves exhibiting the slower residue dissipation was not constant. The first two half-lives for groves 5-7 can be approximated; they are 11 and 32 d, respectively.

The third investigation involved an illness episode in which 11 raisin harvesters developed acute respiratory irritation symptoms when they were exposed to residues of λ-cyhalothrin, propargite and sulfur. Gas chromatography analyses of eight dislodgeable foliar residue (DFR) samples verified mean residues of λ-cyhalothrin ($0.43 \pm 0.10$ µg/cm²), propargite ($0.35 \pm 0.11$ µg/cm²), and sulfur ($0.31 \pm 0.29$ µg/cm²) of the grape leaves. Subsequent investigation confirmed that the λ-cyhalothrin product, which was not registered for use on grapes, was mistakenly mixed and applied 45 d earlier at 35 times the highest legal rate for any crop. The effects of exposure to average λ-cyhalothrin DFR levels of 0.43 µg/cm² have not been previously documented.

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